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Relevance of interfacial viscoelasticity in stability and conformation of biomolecular organizates at air/fluid interface

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ABSTRACT

Soft materials are complex macromolecular systems often exhibiting perplexing non-Newtonian viscoelastic properties, especially when the macromolecules are entangled, crowded or cross-linked. These materials are ubiquitous in the biology, food and pharma industry and have several applications in biotechnology and in the field of biosensors. Based on the length scales, topologies, flexibility and concentration, the systems behave both as liquids (viscous) and solids (elastic). Particularly, for proteins and protein–lipid systems, viscoelasticity is an important parameter because it often relates directly to stability and thermodynamic interactions of the pure biological components as well as their mixtures. Despite the large body of work that is available in solution macro-rheometry, there are still a number of issues that need to be addressed in dealing with proteins at air/fluid interfaces and with protein–polymer or protein–lipid interfaces that often exhibit very low interfacial viscosity values.

Considering the important applications that they have in biopharmaceutical, biotechnological and nutraceutical industries, there is a need for developing methods that meet the following three specific issues: small volume, large dynamic range of shear rates and interfacial properties of different biomolecules. Further, the techniques that are developed should include Newtonian, shear thinning and yielding properties, which are representative of the different solution behaviors typically encountered. The review presented here is a comprehensive account of the rheological properties of different biomolecules at air/fluid and solid/fluid interfaces. It addresses the usefulness of 'viscoelasticity' of the systems at the interfaces analyzed at the molecular level that can be correlated with the microscopic material properties and touches upon some recent techniques in microrheology that are being used to measure the unusually low viscosity values sensitively.

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1. Introduction

Soft materials that are composed of colloidal particles, filamentous polymers and other supra-molecular arrangements of biomolecules or composites of these with polymers have internal structures that show complicated deformations in response to mechanical stress. They exhibit both elastic and viscous responses and are therefore 'viscoelastic'. As a result, the relations between stress and strain are not simply defined by elastic and viscous constants; rather, they are functions of time, direction and extent of deformation. In the design and application of these materials, many open questions remain about how to relate their structure to viscoelastic response. Fig.1 depicts the dynamic response of protein systems (some cross-linked) with the dynamic shear storage moduli measured for different strain amplitudes.

Usually for a flexible polymer, the response would be linear with the G_0 being constant for strain even up to 100%. On the other hand, fibrous proteins would exhibit stiffening behavior on reaching 'gel' state even at medium strain levels. Thus, they show a large increase in shear elastic moduli for a small increase in strain.

In case of proteins and or protein–polymer composites, be it the design of a new biomaterial or new colloidal dispersions for modern drug or nutraceutical delivery systems, one of the bottlenecks has been the thermodynamic instability caused by their large interfacial area. Reducing this instability is therefore crucial to improve the efficacy of the delivery systems, for which an understanding of the behavior of materials at interfaces is required. The stability of these systems is improved by using surfactants, proteins, surface active polymers and amphiphilic compounds. Here, surface viscosity and elasticity usually arise at the fluid interfaces due to adsorption of proteins, polymeric surfactants, low molecular weight surfactants and combination thereof that effectively reduce the interfacial tension [1,2]. In the context of many technological applications in food processing, detergent, cosmetic, pharmaceuticals industry or even mining, specifically in foam and emulsion formation, the dynamic interfacial properties play an important role [3–5]. Specifically, the interfacial viscoelasticity is crucial in determining the efficacy of drug delivery systems, in preparation of formulations through liposomes, in design of new materials, in spray coating, in storage of proteins using freeze drying, Langmuir–Blodgett films, liquid/liquid deposition, two phase flows and mass transfer in industrial preparations, in new biomaterials (Fig. 2). Some of the protein–surfactant systems in the above materials are also commonly found in many biological systems that include blood, motor proteins and wound healing processes in extra cellular matrix systems [6–9].

2. Modeling viscoelastic effects

Dimakopoulos et al. have used a numerical algorithm based on a two-dimensional mixed finite element method to study steady state blood flow in stenotic rigid vessels [6]. Their results showed that viscoelastic effects in blood flow are responsible for steeper decreases of tube and discharge hematocrits as decreasing function of constriction ratio in blood vessels.

Nam and Epureanu studied the effect of varying viscoelasticity on kinesins–molecular motors and have shown that highly viscoelastic fluids have considerable effects on the movement of kinesin and that the high viscosity modifies the relation between the load and the speed of kinesin. Using this model, they predicted the motion of kinesin under time-varying loads [7].

Kim et al. have examined the structural changes and mechanical responses of fibrin networks exposed to compressive loads. They used rheology with simultaneous confocal microscopy and analyzed structural modulations underlying nonlinear viscoelasticity of compressed fibrin networks. Their results showed that repeated compression/decompression enhanced fibrin clot stiffening [8].

Ahmadzadeh et al. applied a microstructural model of the axonal cytoskeleton to elucidate the interaction between microtubules and tau proteins under mechanical loading [9]. Using single molecule force spectroscopic studies, they found that the viscoelastic behavior of tau proteins leads to mechanical breaking of microtubules at high strain rates, and extension of tau allows for reversible sliding of microtubules

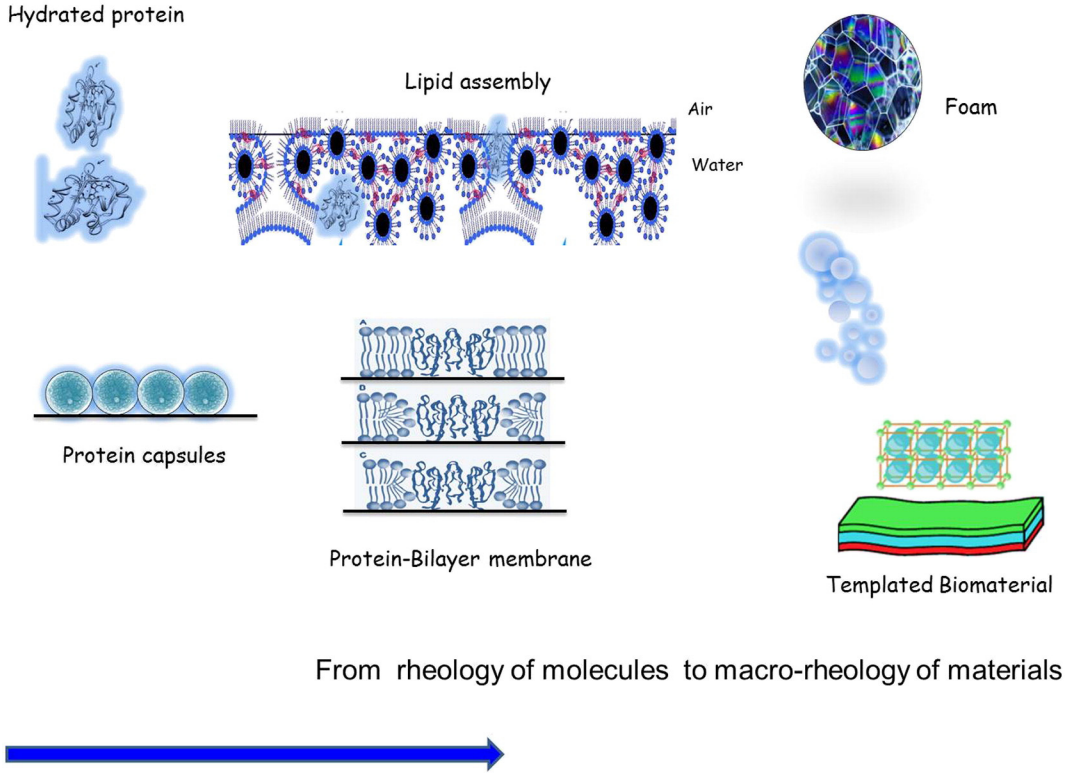

Fig. 1. Dynamic response of protein systems (some cross-linked) presented using change in the dynamic shear storage moduli measured for different strain amplitudes.

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